The surface electronic structure of ferromagnetic Fe(001)

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A thorough investigation of the surface electronic structure of ferromagnetic Fe(100) films, epitaxially grown on single crystal W(100), has been conducted using spin- and angle-resolved photoemission combined with *state-of-the-art* density functional theory slab-computations. The dispersion of the surface state emission close to the Fermi level has been assessed quantitatively. The experimental results are in a good agreement with the calculations. In particular the presence of a minority surface state with d_{xz+yz} character along the $\overline{\Gamma X}$ high-symmetry direction is unambiguously established. Additionally, the calculations predict the existence of an unoccupied surface state localized at $\overline{\Gamma}$. The presence of the related minority interface resonance near the Fermi edge and outside of the surface-Brillouin-zone center $\overline{\Gamma}$ is believed to control the tunneling magneto-resistance in Fe/MgO/Fe(001) for very thin MgO spacers. Our results indirectly confirm these predictions.

FIG. 1: *Minority-spin band structure of Fe(001) along $\overline{\Gamma X}$ calculated using WIEN2k [4] for a 30 atoms slab. Adjacent slabs were separated by 29.8 Å of vacuum. States with more than 20% charge located on the surface atom are emphasized as thicker (red) lines. Only even states with respect to the emission plane are plotted since only these states are accessible in our experimental geometry.*

The nature of the surface states in iron has been an area of intense research spanning several decades. Due to their high localization, surface states of magnetic materials have been closely scrutinized for signs of surface-enhanced electronic correlation, surface-dominated magnetic anisotropies, and surface-enhanced magnetism. Methods of choice to investigate these surface states have been angular-resolved photoemission (ARPES) and its spin-polarized version SP-ARPES, yielding detailed insights into the spin-split electronic structure and the symmetry of the occupied and unoccupied electronic states. In spite of these efforts, the determination of the surface photoemission contribution from a seemingly simple system like ferromagnetic Fe(001) is still somewhat unclear. One of the reasons for this is the interplay of exchange and spin-orbit interaction, which tend to split and mix states particularly in the vicinity of the Fermi level. Another reason for this is the insufficient description of electronic correlations in standard density functional theory approaches.

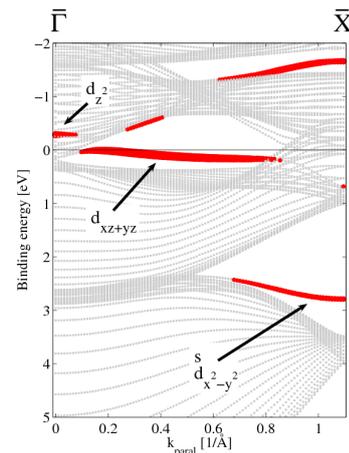The mechanism of tunneling in Fe/MgO/Fe(001) magnetic tunnel junctions (MTJs) may be understood on the basis of band structure theory [1]. In real devices only local atomic order is mandatory, thus textured interfaces are often sufficient, being easier in preparation. Photoemission is a laterally averaging approach, thus capturing what happens *locally* in the real MTJ requires preparation of *long-range* atomically ordered films under UHV.

In this study we examine the Fe(001) surface emission by using SP-ARPES [2]. Although being limited to sampling occupied states – contrary to STM studies – these type of measurements probe the k-dependence and spin-character of the occupied electronic bands directly, together with their energy distribution. Furthermore, by adjusting photon energy and emission angle, several distinct points in the bulk Brillouin zone can be sampled. We compare a new set of experimental data to a state-of-the-art band structure computation. The minority surface state is observed to cross the Fermi level and to become unoccupied in the proximity of $\overline{\Gamma}$ as predicted by the calculations. We also examine the effects of oxygen adsorption on the relative intensities of various features in valence band spectra, which provides an additional evidence whether they are bulk- or surface-related.
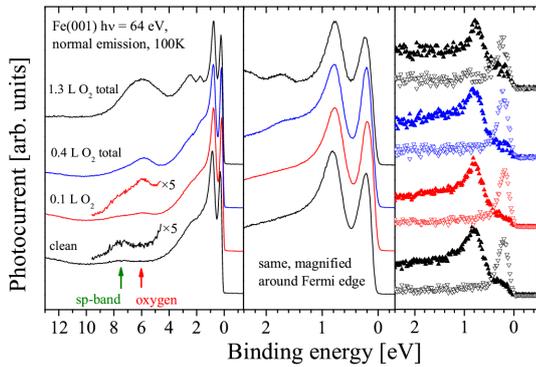
FIG. 2: Oxygen adsorption on Fe(001): normal emission spectra at $h\nu$ = 64 eV. Both spin-integrated (left and center panels) and spin-polarized (right panel) spectra are shown; ▲ - majority states, ▽ - minority states.
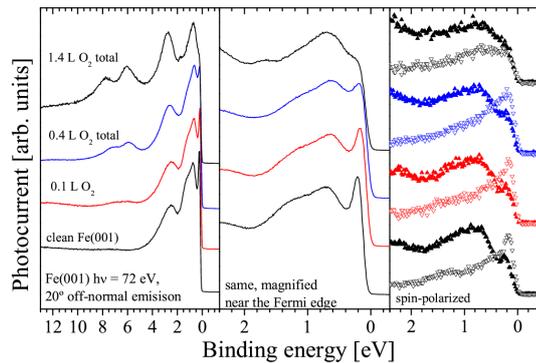

FIG. 3: Oxygen adsorption on Fe(001): 20° off-normal emission spectra at $h\nu$ = 72 eV probing states in the bulk BZ where a strong minority peak appears near the Fermi edge [2]. See caption of Fig. 2 for details.

We have grown relatively thick Fe(001) films (~ 30 Å) on a W(001) single crystal at 100 K at a rate of ~ 8 Å per minute by electron beam evaporation. The base pressure in the chamber was 5×10^{-11} Torr and rose to about 1×10^{-10} Torr during Fe deposition. After deposition the sample was flashed to 400 K to improve crystallinity and subsequently cooled down again to 100 K. Low energy electron diffraction (LEED) observations indicate that above a few monolayers thickness, such a procedure yields epitaxial films with a relaxed bulk-Fe lattice constant.

Figure 2 compiles normal emission data taken at 64 eV photon energy for the clean and oxygen exposed Fe(001) surface. Adopting the usual free-electron-like final-state, bulk states close to the Γ point are excited at this photon energy. In order to test for any surface related emission in these spectra, experiments with carefully controlled oxygen exposures were performed. The region near the Fermi edge is dominated by a strong minority feature, which, however, is not influenced by oxygen exposure. This indicates that the electronic origin of this feature is rather of a bulk than surface nature.

In order to find the occupied surface state described in the calculations above, we have continued the search by mapping the band structure along ΓH in off-normal emission and sharp emission features

near the Fermi edge were observed especially for 67 eV, 12° and 72 eV, 20° (actually both having minority character) [2]. The latter parameters have been chosen for more detailed spin-polarized measurements. Figure 3 presents the results of the oxygen absorption test for the off-normal emission condition. This time the minority emission close to the Fermi level is strongly suppressed by the presence of adsorbed oxygen, clearly indicating its surface origin. It should, however, be noted that in order to completely quench this state relatively high oxygen exposures (1.4 Langmuir) are needed. By contrast, for smaller doses (up to 0.4 L) it remains practically unaffected. Oxygen is known to form epitaxial crystalline overlayers on the Fe(001) surface, and indeed a single broad feature is observed in normal emission, while two distinct features are seen in the spectra 20° off-normal emission, which indicates a dispersive character. Possibly the surface state is pushed above the Fermi level only when a certain fraction of the surface is covered with oxygen.

In summary we have established details of the occupied minority spin surface state in Fe(001). The minority spin surface state of d_{xz+yz} symmetry lies in the gap of the projected bulk spin-down band structure of even symmetry. It is occupied for the major part of the $\overline{\Gamma X}$ distance and disappears both near $\overline{\Gamma}$ and near $\overline{X}$. Around $\overline{\Gamma}$ there exists another unoccupied surface state of d_{z^2} character, which was previously observed [3]. Adsorption of an ordered monolayer of oxygen quenches the surface state, however, without removing the surface ferromagnetism [5]. Our results provide new important information regarding the details of the electronic states which are believed to play an essential part in the functionality of Fe/MgO/Fe(001)-based MTJs. Further off-normal emission experimental results and detailed theoretical calculations are needed to establish how MgO overlayers influence the minority surface state of Fe(001).

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